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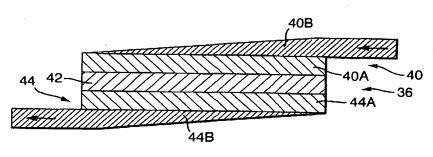
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(54) Title: A HIGH-TEMPERATURE FUEL CELL MODULE



(57) Abstract: A solid oxide fuel cell module (30) comprises a plurality of fuel cells (36). Each fuel cell (36) comprises a first electrode (40), an electrolyte (42) and a second electrode (44). A plurality of interconnectors (38) are arranged to electrically connect the fuel cells (36) in electrical series. Each interconnector (38) electrically connects a first electrode (40) of one fuel cell (36) to a second electrode (44) of an adjacent fuel cell (36). The first electrode (40) comprises a first layer (40A) on the electrolyte (42) to optimise the electrochemical activity at the electrolyte (42) and a second layer (40B) on the first layer (40A) to provide electronic conduction perpendicular to the layers (40, 42, 44) of the fuel cell (36). The second layer (40B) is arranged such that electronic conduction perpendicular to the layers (40, 42, 44) of the fuel cell (36) is different at different positions in the second layer (40B). This can be achieved through a different thickness, different composition or different geometry of the second layer (40B) at different positions.

#### A HIGH-TEMPERATURE FUEL CELL MODULE

The present invention relates to a fuel cell module, in particular to a solid oxide fuel cell module.

Solid oxide fuel cell modules comprising a plurality of solid oxide fuel cells connected in electrical series are known. The solid oxide fuel cells are connected in series by interconnectors.

Solid oxide fuel cells comprising functionally graded anode electrodes and cathode electrodes are known. The functionally graded anode electrodes and cathode electrodes generally comprise a first layer on the electrolyte and a second layer on the first layer. The first layer is arranged to optimise the electrochemical activity at the electrolyte and the second layer is arranged to provide electronic conduction perpendicular to the layers of the solid oxide fuel cells to allow current to flow from one solid oxide fuel cell to an adjacent solid oxide fuel cell via an interconnector. The second layers provide uniform current collection across the solid oxide fuel cells.

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Accordingly the present invention seeks to provide a novel fuel cell module.

Accordingly the present invention provides a solid oxide fuel cell module comprising a plurality of fuel cells, each fuel cell comprising a first electrode, electrolyte 25 second electrode, a plurality and a interconnectors being arranged to electrically connect the fuel cells in electrical series, each interconnector electrically connecting a first electrode of one fuel cell to a second electrode of an adjacent fuel cell, the first electrode comprising a first layer on the electrolyte to optimise the electrochemical activity at the electrolyte and a second layer on the first layer to provide electronic conduction perpendicular to the layers of the fuel cell, layer being arranged such that electronic conduction perpendicular to the layers of the fuel cell is different at different positions in the second layer.

Preferably the second electrode comprising a first layer on the electrolyte to optimise the electrochemical activity at the electrolyte and a second layer on the first layer to provide electronic conduction perpendicular to the layers of the solid oxide fuel cell, the second layer being arranged such that electronic conduction perpendicular to the layers of the fuel cell is different at different positions in the second layer.

The second layer may have a different thickness at 10 different positions such that electronic conduction perpendicular to the layers of the fuel cell is different at different positions in the second layer.

Alternatively the second layer has a different composition at different positions such that electronic conduction perpendicular to the layers of the fuel cell is different at different positions in the second layer.

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Preferably the second layer has a different geometry at different positions such that electronic conduction perpendicular to the layers of the fuel cell is different at different positions in the second layer.

Preferably the second layer comprises a mesh having a plurality of holes, the holes have a different cross-sectional areas at different positions such that electronic conduction perpendicular to the layers of the fuel cell is different at different positions in the second layer.

Preferably the second layer is arranged such that at a first position near to the interconnector the electronic conduction perpendicular to the layers of the fuel cell is greater than the electronic conduction perpendicular to the layers of the fuel cell at a second position remote from the interconnector.

Preferably the second layer is arranged such that the electronic conduction perpendicular to the layers of the fuel cell progressively decreases between the first position and the second position.

Preferably the fuel cell module comprising a hollow support member, the fuel cells being spaced apart on at least one surface of the hollow support member.

The present invention also provides a solid oxide fuel cell comprising a first electrode, an electrolyte and a second electrode, the first electrode comprising a first layer on the electrolyte to optimise the electrochemical activity at the electrolyte and a second layer on the first layer to provide electronic conduction perpendicular to the layers of the fuel cell, the second layer being arranged such that electronic conduction perpendicular to the layers of the fuel cell is different at different positions in the second layer.

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Preferably the second electrode comprising a first layer on the electrolyte to optimise the electrochemical activity at the electrolyte and a second layer on the first layer to provide electronic conduction perpendicular to the layers of the fuel cell, the second layer being arranged such that electronic conduction perpendicular to the layers 20 of the fuel cell is different at different positions in the second layer.

The second layer may have a different thickness at positions that electronic conduction such perpendicular to the layers of the fuel cell is different at different positions in the second layer.

Alternatively the second layer has a different composition at different positions such that electronic conduction perpendicular to the layers of the fuel cell is different at different positions in the second layer.

30 Preferably the second layer has a different geometry different positions such that electronic conduction perpendicular to the layers of the fuel cell is different at different positions in the second layer.

Preferably the second layer comprises a mesh having a plurality of holes, the holes have a different crosssectional areas at different positions such that electronic conduction perpendicular to the layers of the fuel cell is different at different positions in the second layer.

Preferably the second layer is arranged such that at a first position the electronic conduction perpendicular to the layers of the fuel cell is greater than the electronic conduction perpendicular to the layers of the fuel cell at a second position remote from the first position.

Preferably the second layer is arranged such that the electronic conduction perpendicular to the layers of the fuel cell progressively decreases between the first position and the second position.

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Preferably the fuel cell comprises a solid oxide fuel cell.

The present invention will be more fully described by 15 way of example with reference to the accompanying drawings in which:-

Figure 1 is a schematic cross-section through a prior art solid oxide fuel cell module.

Figure 2 is an enlarged schematic cross-section 20 through a single solid oxide fuel cell shown in figure 1.

Figure 3 is a schematic cross-section through a solid oxide fuel cell module according to the present invention.

Figure 4 is an enlarged schematic cross-section through a single solid oxide fuel cell according to the present invention shown in figure 3.

Figure 5 is an enlarged schematic cross-section through a further solid oxide fuel cell according to the present invention shown in figure 3.

Figure 6 is an enlarged schematic cross-section 30 through another solid oxide fuel cell according to the present invention shown in figure 3.

Figure 7 is a view in the direction of arrow A in figure 4.

A prior art solid oxide fuel cell module 10 is shown in figures 1 and 2. The solid oxide fuel cell module 10 comprises a hollow support member 12 and a plurality of

solid oxide fuel cells 16 spaced apart longitudinally on at least one flat surface 14 of the hollow support member 12. The solid oxide fuel cells are electrically connected in series by a plurality of interconnectors 18. Each solid oxide fuel cell 16 comprises a first electrode 20, a cathode electrode, an electrolyte 22 and a second electrode 24, an anode electrode. The second electrode 24 is arranged on the surface 14 of the hollow support member 12, the electrolyte 22 is arranged on the second electrode 24 and the first electrode 20 is arranged on the electrolyte 22.

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The first electrode 20 and the second electrode 24 are functionally graded, as shown more clearly in figure 2. The functionally graded first electrode comprises a first layer 20A on, or adjacent, the electrolyte 22 and a second 15 layer 20B on, or adjacent, the first layer 20A. functionally graded second electrode 24 comprises a first layer 24A under, or adjacent, the electrolyte 22 and a second layer 24B under, or adjacent, the first layer 24A. The first layers 20A, 24A are arranged to optimise the 20 electrochemical activity at the electrolyte 22 and the second layers 20B, 24B are arranged to provide electronic conduction perpendicular to the layers 20, 22, 24 of the solid oxide fuel cells 16 to allow current to flow from one solid oxide fuel cell 16 to an adjacent solid oxide fuel cell 16 via a respective interconnector 18. The second layers 20B, 24B provide uniform current collection across the solid oxide fuel cells 16.

A solid oxide fuel cell module 30 according to the present invention is shown in figures 3 and 4. The solid oxide fuel cell module 30 comprises a hollow support member 32 and a plurality of solid oxide fuel cells 36 spaced apart longitudinally on at least one flat surface 34 of the hollow support member 32. The solid oxide fuel cells are electrically connected in series 35 by a plurality οf interconnectors 38. Each solid oxide fuel cell 36

comprises a first electrode 40, cathode electrode, an electrolyte 42 and a second electrode 44, anode electrode. The second electrode 44 is arranged on the surface 34 of the hollow support member 32, the electrolyte 42 is arranged on the second electrode 44 and the first electrode 40 is arranged on the electrolyte 42.

The first electrode 40 and the second electrode 44 are functionally graded, as shown more clearly in figure 4. The functionally graded first electrode comprises a first layer 40A on, or adjacent, the electrolyte 42 and a second layer 40B on, or adjacent, the first layer 40A. functionally graded second electrode 44 comprises a first layer 44A under, or adjacent, the electrolyte 42 and a second layer 44B under, or adjacent, the first layer 44A. The first layers 40A, 44A are arranged to optimise the 15 electrochemical activity at the electrolyte 42 and second layers 40B, 44B are arranged to provide electronic conduction perpendicular to the layers 40, 42, 44 of the solid oxide fuel cells 36 to allow current to flow from one 20 solid oxide fuel cell 36 to an adjacent solid oxide fuel cell 36 via a respective interconnector 38.

The second layers 40B, 44B the second layer being arranged to provide differential current collection across the solid oxide fuel cells 36. The second layers 40B, 44B are arranged such that electronic conduction perpendicular to the layers 40, 42, 44 of the solid oxide fuel cells 16 is different at different positions in the second layers 40B, 44B.

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In this example the second layers 40B, 44B have a different thickness at different positions such that the electronic conduction perpendicular to the layers of the solid oxide fuel cells 16 is different at different positions in the second layers 40B, 44B.

In particular it is seen that the thickness of the second layer 40B of the first electrode 40 is greatest at a first end of the first electrode 40 nearest the

interconnector 38 and the thickness of the second layer 40B of the first electrode 40 is least at the end of the first electrode 40 remote from the interconnector 18. The thickness of the second layer 40B gradually, continuously, decreases, or tapers, from the first end to the second end of the first electrode 40.

Similarly the thickness of the second layer 44B of the second electrode 44 is greatest at the end of the second electrode 44 nearest the interconnector 38 and the thickness of the second layer 44B of the second electrode 44 is least at the end of the second electrode 44 remote from the interconnector 38. The thickness of the second layer 44B gradually, continuously, decreases, or tapers, from the first end to the second end of the second electrode 44.

Alternatively it may be possible to decrease the thickness of the second layers 40B, 44B in steps.

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The losses associated with the second layers 40B, 44B are ohmic losses related to the current and the resistance and are simply expressed as I<sup>2</sup>R. In this arrangement the conductivity of the second layers 40B, 44B is related to the cross-sectional area of the second layers 40B, 44B. This arrangement reduces the amount of material used in the second layers 40B, 44B and reduces the costs of the material used in manufacturing the solid oxide fuel cells 16.

The second layers 40B, 44B comprise at least one of palladium, platinum, silver, gold, nickel, copper, cobalt, chromium, iron or ruthenium or an alloy of two or more of these elements.

The thickness of the second layers 40B, 44B may be 100 micrometers at the first end and 1 micrometer at the second end.

A further solid oxide fuel cell module 50 according to the present invention is shown in figures 3 and 5. The

solid oxide fuel cell module 50 is similar to that shown in figures 3 and 4.

In this example the second layers 40B, 44B have a different compositions at different positions such that the electronic conduction perpendicular to the layers of the solid oxide fuel cells 16 is different at different positions in the second layers 40B, 44B. The compositions of the second layers 40B, 44B are varied by varying the proportions of a high conductivity material and a low conductivity material.

In particular the composition of the second layer 40B of the first electrode 40 comprises a greater proportion of a high conductivity material at a first end of the first electrode 40 nearest the interconnector 38 and composition of the second layer 40B of the first electrode 40 comprises a lesser proportion of the high conductivity material at the second end of the first electrode 40 remote from the interconnector 18. The proportion of conductivity material in the second layer 40B gradually, continuously, reduces from the first end to the second end of the first electrode 40.

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Similarly the composition of the second layer 44B of the second electrode 44 is comprises a greater proportion of the high conductivity material at a first end of the second electrode 44 nearest the interconnector 38 and the composition of the second layer 44B of the second electrode 44 comprises a lesser proportion of the high conductivity material at the second end of the second electrode 44 remote from the interconnector 38. The proportion of high conductivity material in the second layer 44B gradually, continuously, reduces, from the first end to the second end of the second electrode 44.

Alternatively the proportion of high conductivity material in the second layer 40B, 44B may reduce in steps rather than continuously.

It is essential to control the microstructure of the second layers 40B, 44B to maintain a constant porosity to allow the oxidant/fuel to reach the first layers 40A, 44A of the first electrode 40 and second electrode 44.

of palladium, platinum, silver, gold, nickel, copper, cobalt, chromium, iron or ruthenium or an alloy of two or more of these elements. The low conductivity material comprises lanthanum manganite, lanthanum cobaltite, a cermet or an amorphous metal in a glass phase. The cermet comprises at least one of palladium, platinum, silver, gold, nickel, copper, cobalt, chromium, iron or ruthenium or an alloy of two or more of these elements in zirconia or ceria.

As an example the composition at the first end of the second layers 40B, 44B is 100% high conductivity material.

A further solid oxide fuel cell module 50 according to the present invention is shown in figures 3, 6 and 7. The solid oxide fuel cell module 50 is similar to that shown in 20 figures 3 and 4.

In this example the second layers 40B, 44B have a different geometry at different positions such that the electronic conduction perpendicular to the layers of the solid oxide fuel cells 16 is different at different positions in the second layers 40B, 44B.

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In particular it is seen that the cross-sectional area of the second layer 40B in contact with the first layer 40A is greatest at a first end of the first electrode 40 nearest the interconnector 38 and the cross-sectional area of the second layer 40B in contact with the first layer 40A is least at the end of the first electrode 40 remote from the interconnector 18. The cross-sectional area of the second layer 40B in contact with the first layer 40A gradually, continuously, decreases, from the first end to the second end of the first electrode 40.

Similarly the cross-sectional area of the second layer 44B in contact with the first layer 44A is greatest at the end of the second electrode 44 nearest the interconnector 38 and the cross-sectional area of the second layer 44B in 5 contact with the first layer 44A is least at the end of the second electrode 44 remote from the interconnector 38. cross-sectional area of the second layer 44B in contact with the first layer 44A gradually, continuously, decreases, from the first end to the second end of the second electrode 44.

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The second layers 40B, 44B are meshes, that is the second layers 40B, 44B comprises a plurality of holes 50 around which are lands 52. The number of holes per unit area remains constant but the cross-sectional area of the holes gradually increases from the first end to the second end of the second layers 40B, 44B. Alternatively the cross-sectional area of the holes increases in steps. Alternatively the cross-sectional area of the holes remains constant, but the number of holes gradually increases from 20 the first end to the second end of the second layers 40B, These meshes are preferably produced by screenprinting, but other suitable methods may be used.

The pitch dimension between holes is between 0.05mm and 5mm, the diameter of the holes is between 0.05mm and 5mm and the lands have a dimension between 0.05mm 25 0.5mm.

The present invention has been described by way of reference to simple schematic representations of a solid oxide fuel cell module. In practice each of the first electrode, second electrode and electrolyte of each solid oxide fuel cell may comprise one or more layers tailored for particular properties. Seals may be provided to prevent leakage of reactants from the first electrode and second electrode of the solid oxide fuel cells. present invention is applicable to these practical solid oxide fuel cell modules.

#### Claims: -

- A solid oxide fuel cell module (30) comprising a plurality of solid oxide fuel cells (36), each fuel cell (36) comprising a first electrode (40), an electrolyte (42) and a second electrode (44), a plurality of interconnectors (38) being arranged to electrically connect the fuel cells (36)electrical series, each interconnector electrically connecting a first electrode (40) of one fuel cell (36) to a second electrode (44) of an adjacent fuel cell (36), the first electrode (40) comprising a first on the electrolyte (42) to optimise layer (40A) electrochemical activity at the electrolyte (42) and a second layer (40B) on the first layer (40A) to provide electronic conduction perpendicular to the lavers (40,42,44) of the fuel cell (36), characterised in that the 15 second layer (40B) being arranged such that electronic conduction perpendicular (40,42,44) to the layers of the fuel cell (36) is different at different positions in the second layer (40B).
- A solid oxide fuel cell module as claimed in claim 1 20 wherein the second electrode (44) comprising a first layer (44A)on the electrolyte (42) to optimise electrochemical activity at the electrolyte (42) and a second layer (44B) on the first layer (44A) to provide 25 electronic conduction perpendicular to the (40,42,44) of the fuel cell (36), the second layer (44B)arranged such that electronic conduction perpendicular to the layers (40,42,44) of the fuel cell (36) is different at different positions in the second 30 layer.
  - 3. A solid oxide fuel cell module as claimed in claim 1 or claim 2 wherein the second layer (40A,40B) having a different thickness at different positions such that electronic conduction perpendicular to the layers (40,42,44) of the fuel cell (36) is different at different positions in the second layer (40B,44B).

- 4. A solid oxide fuel cell module as claimed in claim 1 or claim 2 wherein the second layer (40B,44B) having a different composition at different positions such that electronic conduction perpendicular to the layers (40,42,44) of the fuel cell (36) is different at different positions in the second layer (40B,44B).
  - 5. A solid oxide fuel cell module as claimed in claim 1 or claim 2 wherein the second layer (40B,44B) having a different geometry at different positions such that electronic conduction perpendicular to the layers (40,42,44) of the fuel cell (36) is different at different positions in the second layer (40B,44B).

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- 6. A solid oxide fuel cell module as claimed in claim 5 wherein the second layer (40B,44B) comprises a mesh having a plurality of holes (50), the holes (50) have different cross-sectional areas at different positions such that electronic conduction perpendicular to the layers (40,42,44) of the fuel cell (36) is different at different positions in the second layer (40B,44B).
- 7. A solid oxide fuel cell module as claimed in any of claims 1 to 6 wherein the second layer (40B,44B) is arranged such that at a first position near to the interconnector (38) the electronic conduction perpendicular to the layers (40,42,44) of the fuel cell (36) is greater than the electronic conduction perpendicular to the layers (40,42,44) of the fuel cell (36) at a second position remote from the interconnector (38).
- 8. A solid oxide fuel cell module as claimed in claim 7 wherein the second layer (40B,44B) is arranged such that 30 the electronic conduction perpendicular to the layers (40,42,14) of the fuel cell (36) progressively decreases between the first position and the second position.
  - 9. A solid oxide fuel cell module as claimed in any of claims 1 to 8 wherein the fuel cell module (30) comprising a hollow support member (32), the fuel cells (36) being

spaced apart on at least one surface (34) of the hollow support member (32).

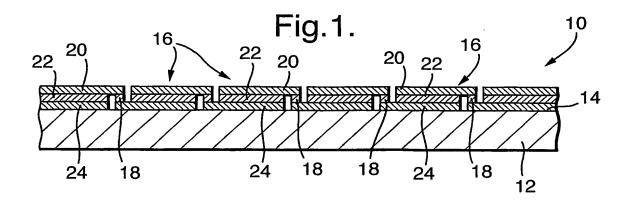
- A solid oxide fuel cell (36) comprising a first electrode (40), an electrolyte (42) and a second electrode (44), the first electrode (40) comprising a first layer (40A) on the electrolyte (42) to optimise electrochemical activity at the electrolyte (42) second layer (40B) on the first layer (40A) to provide electronic conduction perpendicular to the (40,42,44) of the fuel cell (36), characterised in that the 10 second layer (40B) being arranged such that electronic conduction perpendicular to the layers (40,42,44) of the fuel cell (36) is different at different positions in the second layer (40B).
- A solid oxide fuel cell as claimed in claim 10 wherein 15 the second electrode (44) comprising a first layer (44A) on the electrolyte (42) to optimise the electrochemical activity at the electrolyte (42) and a second layer (44B) on the first layer (44A) to provide electronic conduction perpendicular to the layers (40,42,44) of the fuel cell 20 (36), the second layer (44B) being arranged such that electronic conduction perpendicular to the (40,42,44) of the fuel cell (36) is different at different positions in the second layer (44B).
- 25 12. A solid oxide fuel cell as claimed in claim 10 or claim 11 wherein the second layer (40B,44B) has a different thickness at different positions such that electronic conduction perpendicular to the layers (40,42,44) of the fuel cell (36) is different at different positions in the second layer (40B,44B).
  - 13. A solid oxide fuel cell as claimed in claim 10 or claim 11 wherein the second layer (40B,44B) has a different composition at different positions such that electronic conduction perpendicular to the layers (40,42,44) of the fuel cell (36) is different at different positions in the second layer (40B,44B).

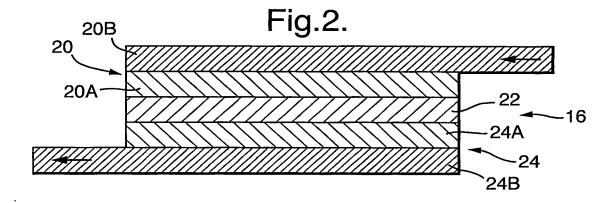
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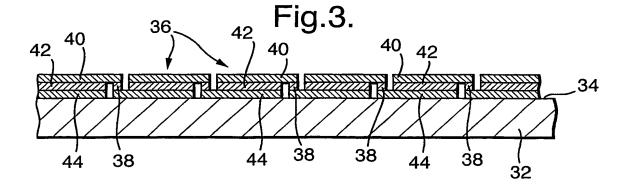
- 14. A solid oxide fuel cell as claimed in claim 10 or claim 11 wherein the second layer (40B,44B) has a different geometry at different positions such that electronic conduction perpendicular to the layers (40,42,44) of the fuel cell (36) is different at different positions in the second layer (40B,44B).
- 15. A solid oxide fuel cell as claimed in claim 14 wherein the second layer (40B,44B) comprises a mesh having a plurality of holes (50), the holes (50) have different cross-sectional areas at different positions such that electronic conduction perpendicular to the layers (40,42,44) of the fuel cell (36) is different at different positions in the second layer (40B,44B).

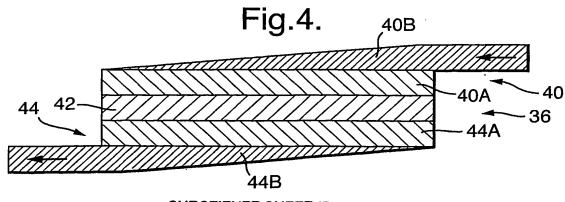
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- 16. A solid oxide fuel cell as claimed in claim 15 wherein the second layer (40B,44B) is arranged such that at a first position the electronic conduction perpendicular to the layers (40,42,44) of the fuel cell (36) is greater than the electronic conduction perpendicular to the layers (40,42,44) of the fuel cell (36) at a second position remote from the first position.
- 17. A solid oxide fuel cell as claimed in claim 16 wherein the second layer (40B,44B) is arranged such that the electronic conduction perpendicular to the layers (40,42,44) of the fuel cell (36) progressively decreases between the first position and the second position.









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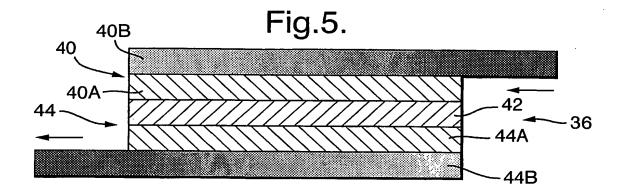
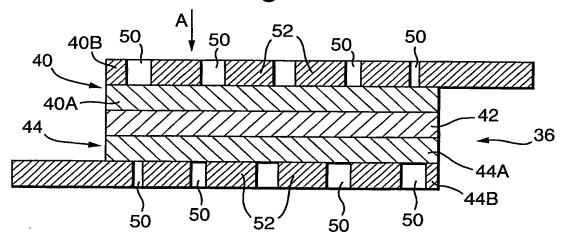
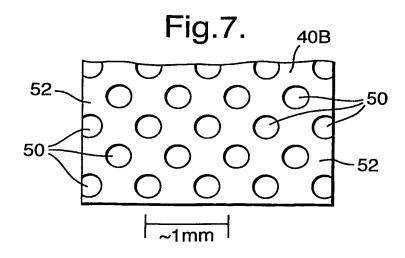


Fig.6.





#### INTERNATIONAL SEARCH REPORT

Intern al Application No PCT/GB 03/00272

A. CLASSIFICATION OF SUBJECT MATTER IPC 7 H01M8/12 H01M H01M8/02 H01M8/24 According to International Patent Classification (IPC) or to both national classification and IPC B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) IPC 7 HO1M Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Electronic data base consulted during the international search (name of data base and, where practical, search terms used) EPO-Internal, PAJ, WPI Data C. DOCUMENTS CONSIDERED TO BE RELEVANT Category ° Citation of document, with indication, where appropriate, of the relevant passages Relevant to claim No. X WO 01 24300 A (FOGER KARL ; JAFFREY DONALD 1-5,9-14 (AU); CERAMIC FUEL CELLS LTD (AU)) 5 April 2001 (2001-04-05) page 10, line 18 -page 11, line 27 figures 1,2 X US 4 699 852 A (YOKOYAMA JIRO ET AL) 1-3, 13 October 1987 (1987-10-13) 7-12,16, column 5-6 figures 1,2 X WO 01 39311 A (US ENERGY ; MICHAEL A COBB & COMPANY (US)) 31 May 2001 (2001-05-31) 7-12,16, page 11, line 28 -page 12, line 22 figure 6 -/--X Further documents are listed in the continuation of box C. Patent family members are listed in annex. Special categories of cited documents: "T" later document published after the international filing date or priority date and not in conflict with the application but "A" document defining the general state of the art which is not cited to understand the principle or theory underlying the considered to be of particular relevance Invention .Е. earlier document but published on or after the international "X" document of particular relevance; the claimed invention filing date cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such document. "O" document referring to an oral disclosure, use, exhibition or other means ments, such combination being obvious to a person skilled \*P\* document published prior to the International filing date but later than the priority date claimed in the art. "&" document member of the same patent family Date of the actual completion of the international search Date of mailing of the International search report 22 May 2003 02/06/2003 Name and mailing address of the ISA Authorized officer European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016 Jacquinot, P

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